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Slow Decreasing Tendency of Fine Particles Compared to Coarse Particles Associated with Recent Hot Summers in Seoul, Korea

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ABSTRACT

In Seoul, South Korea, particulate matters (PMs) significantly decreased for the period 2005–2015 in concert with the implementation of air pollution reduction policies. This study reveals that PM with a diameter smaller than 2.5 µm ($PM_{2.5}$) has a slower decreasing tendency than PM in the 2.5–10-µm range ($PM_{2.5\text{-}10}$) during summer and that this discrepancy is attributable to the large increase in the summer surface air temperature for the analysis period (0.13°C year^{-1}). During the daytime, especially in the afternoon when the hourly surface air temperature and its increasing rate are high, the difference between the decreasing rates of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ is pronounced. The slower decrease in $PM_{2.5}$ compared to $PM_{2.5\text{-}10}$ likely results from the secondary $PM_{2.5}$ formation being accelerated by the increase in the surface air temperature. Other atmospheric variables that can affect concentrations of PMs, such as insolation, relative humidity, precipitation, wind speed, and sea-level pressure, do not show a meaningful relationship with the discrepancy in the decreasing tendency between the two PMs. Our finding emphasizes the necessity of continuous monitoring and analysis of long-term variability in concentrations of PMs and related meteorological conditions, and the independent establishment of reduction policies for $PM_{2.5}$ and $PM_{2.5\text{-}10}$ to prepare for anthropogenic climate change and the subsequent air quality change.

Keywords: $PM_{2.5}$; $PM_{2.5\text{-}10}$; Surface air temperature; Secondary formation; Urban area.

INTRODUCTION

Particulate matters with a diameter smaller than 2.5 µm ($PM_{2.5}$) and in the 2.5–10 µm range ($PM_{2.5\text{-}10}$) have negative effects on human health (Wilson and Suh, 1997). Because of the distinct differences in the physical and chemical properties of $PM_{2.5}$ and $PM_{2.5\text{-}10}$, their adverse impacts on human health are diverse. Many epidemiologic studies have suggested that short-term exposure to $PM_{2.5}$ causes respiratory and cardiovascular problems, while similar exposure to $PM_{2.5\text{-}10}$ causes pulmonary diseases and asthma (Brunekreef and Forsberg, 2005; Perez *et al.*, 2009). For long-term exposures to PMs, $PM_{2.5}$ is particularly related to mortality from cardiovascular disease; the effects of $PM_{2.5\text{-}10}$ exposure on mortality are not clear because of the limited number of studies and limitations in exposure assessment (Zhang *et al.*, 2011; Hoek *et al.*, 2013). In order to regulate ambient air quality to reduce adverse impacts of PMs on human health, the Ministry of Environment of South Korea has established National Environmental Comprehensive Plans and implemented nationwide clean-up activities in early 2000s, such as the replacement of diesel-powered buses with natural gas buses, upgrading fuel quality, and regulating air pollutant emissions from factory chimneys (Kim and Shon, 2011a, b). Up until 2015, most reduction policies were designed for PM_{10} concentrations; actions to control $PM_{2.5}$ separately have just begun as a result of the 4th National Environmental Comprehensive Plan for the period 2016–2035 (The Ministry of Environment, 2016).

Studies on the physical and chemical properties of PMs have been conducted for use in policy-making regarding regulation of air pollutant emissions, and eventually, to reduce PMs concentrations below the World Health Organization guidelines—10 µg m^{-3} (25 µg m^{-3}) and 20 µg m^{-3} (50 µg m^{-3}) for the annual (24-hour) average for $PM_{2.5}$ and PM_{10}, respectively (World Health Organization, 2006). In general, $PM_{2.5}$ is primarily formed by fossil fuel

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combustion in vehicles, industrial complexes, and local heating, with the secondary formations formed by chemical reactions; most $PM_{2.5\text{-}10}$ comes from non-exhaust traffic particles, such as tire and brake dust, and wind-blown mineral dust (Castanho and Artaxo, 2001; Cheng *et al.*, 2015). Thus, anthropogenic emissions and the secondary formations tend to increase the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ as a result of larger increases in $PM_{2.5}$ (Amodio *et al.*, 2012; Li *et al.*, 2015), while long-range transport of natural dust (e.g., Asian dust) tends to decrease the ratio by increasing $PM_{2.5\text{-}10}$ (Fu *et al.*, 2010; Zhao *et al.*, 2011). Other properties of PMs, such as sources, compositions, and temporal changes, are varied depending on regions. In terms of $PM_{2.5}$ in Seoul, South Korea, previous studies have reported chemical composition (Park *et al.*, 2002; Kim *et al.*, 2006; Kim *et al.*, 2007), source apportionment (Park and Kim, 2005; Heo *et al.*, 2009), source region (Kang *et al.*, 2006; Han *et al.*, 2008), and temporal variation (Lee *et al.*, 1999; Ghim *et al.*, 2015; Vellingiri *et al.*, 2015). However, most of these studies were based on few long-term monitoring programs for $PM_{2.5}$. For a long-term perspective, it is necessary to study changing proportions and trends of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ in order to enable regulation of their ambient concentrations separately and effectively. Furthermore, meteorological effects on $PM_{2.5}$ and $PM_{2.5\text{-}10}$ need to be analyzed, considering that long-term changes in atmospheric conditions can induce long-term changes in concentrations of PMs.

Many studies have examined the relationship between $PM_{2.5}$ and meteorological variables that affect the primary and secondary formations of $PM_{2.5}$ (Wang *et al.*, 2005; Kim *et al.*, 2007; Theodosi *et al.*, 2011; Barmpadimos *et al.*, 2012; Grivas *et al.*, 2012; Kassomenos *et al.*, 2014). Barmpadimos *et al.* (2011, 2012) showed that $PM_{2.5}$ concentration is negatively correlated with wind speed and precipitation because of the dilution effect and the scavenging effect, respectively. They also indicated that surface air temperature is positively correlated with $PM_{2.5\text{-}10}$ because of dust resuspension in dry conditions, but there is the diverse correlation with $PM_{2.5}$ depending on the season: a negative correlation in winter due to emissions from space heating and a positive correlation in summer due to the secondary formations. Among meteorological variables, surface air temperature is a critical parameter that must be considered in examining the variations of $PM_{2.5}$ concentration, given that it exerts different effects on the primary and secondary $PM_{2.5}$ formations depending on the season. In winter, larger emissions from space heating by wood or fossil fuel burning due to lower air temperature increase primary aerosol concentrations (Dan *et al.*, 2004; Szidat *et al.*, 2007). In summer, high surface air temperature coincident with a large amount of solar radiation can induce the secondary formations of $PM_{2.5}$. Wang *et al.* (2005) suggested that the possibility of photochemical formations of secondary particles such as sulfate and nitrate—water-soluble ions containing the SO_4^{2-} and NO_3^- group—is at its maximum in summer mainly because of high surface air temperature. Kim *et al.* (2007) and Wang *et al.* (2016) suggested that sulfate, nitrate, and ammonium are dominant components of $PM_{2.5}$, especially in summer, accounting for 57% and 47% in Seoul and central China, respectively. Based on the relationship between $PM_{2.5}$ and surface air temperature, it can be hypothesized that, in a long-term perspective, changing surface air temperature affects $PM_{2.5}$ concentration due to the secondary formations and, as a result, induces different changes of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations.

This study examines the trends in $PM_{2.5}$ and $PM_{2.5\text{-}10}$, their ratios, and the effects of surface air temperature on $PM_{2.5}$ concentration. We focus on the differences in the changing rates of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ and the relationship between the changing rates of PMs and surface air temperature. The target area is Seoul, a megacity in South Korea, which is characterized by a high population density of about 17,000 people km^{-2} and heavy traffic of around 3 million vehicles (Korea National Statistical Office, 2006); thus, people have suffered from severe anthropogenic air pollution problems. We first analyze the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ for the period 2005–2015 to investigate the changes in the ratio. Then we examine the difference in the trends of $PM_{2.5}$, $PM_{2.5\text{-}10}$, and surface air temperature for the period. Data are described below. It is followed by analyses of the changes in the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ and trends of $PM_{2.5}$, $PM_{2.5\text{-}10}$, and surface air temperature. Summary follows the results and discussion section.

DATA AND METHOD

Hourly concentrations of $PM_{2.5}$ and PM_{10} were provided by the Seoul Metropolitan Government Research Institute of Public Health and Environment (http://www.open.go.kr). These data have been used recently in several researches (Lee, 2014; Kim *et al.*, 2015; Lee *et al.*, 2015; Kim *et al.*, 2016; Han *et al.*, 2017; Lee *et al.*, 2017). Hourly concentrations of two PM values were measured by using Thermo FH62C-14 (USA) and Kimoto PM-711 (Japan). The accuracy, the precision, and the detection limit of Thermo are ± 5%, ± 2 µg m^{-3}, and 1–10,000 µg m^{-3}, respectively; those of Kimoto are ± 1%, ± 2 µg m^{-3}, and 0.5–5,000 µg m^{-3}, respectively. Mass concentrations of PMs are measured by using the beta-ray absorption method (Kim and Kim, 2003). The measurement error in this method is known to be 10%, which can be caused by particle-containing moisture (Chang and Tsai, 2003; Jung *et al.*, 2007). Measurements of $PM_{2.5}$ and PM_{10} began in 2001 and 1996, respectively. The 25 observation sites are evenly distributed throughout Seoul (Fig. S1), since one site is located per ward (*gu* in Korean), except for Guro-gu and Songpa-gu where two sites for PM_{10} were located until 2008; in each ward, the two concentration values at two sites were averaged. Correlation coefficients between $PM_{2.5}$ (PM_{10}) concentrations in each ward and the others are 0.76–0.95 (0.90–0.97) for the period targeted below. Valid hourly concentrations were calculated with data from at least 50% of sites (13 and more out of 25); and valid daily mean concentrations were calculated with data from at least 50% of sites (13 and more out of 25) and of hourly measurements (12 and more out of 24); otherwise, we omitted the values. $PM_{2.5\text{-}10}$ concentration was obtained by subtracting $PM_{2.5}$ concentration from PM_{10}

concentration, and when either $PM_{2.5}$ or PM_{10} concentration is not valid, $PM_{2.5\text{-}10}$ concentration was omitted. We targeted the period 2005–2015 because of (a) small number of sites for $PM_{2.5}$ until 2004 (less than 13 sites) and (b) abrupt increase in annual mean PM concentration in 2016 (Table S1) —the rise of annual mean $PM_{2.5}$ concentration in 2016 compared to that in 2015 (3.73 µg m^{-3}) is greater than the standard deviation of $PM_{2.5}$ concentration from 2005 to 2016 (2.32 µg m^{-3}). The number of invalid hourly (daily) $PM_{2.5}$ is 1000 (34) and that of invalid hourly (daily) $PM_{2.5\text{-}10}$ is 1002 (34) for the period 2005–2015. To focus on PMs from anthropogenic emissions, we excluded Asian dust days for 2005–2015 using Asian dust events data from the Korea Meteorological Administration (KMA) (http://www.kma.go.kr/weather/asiandust/observday.jsp).

Hourly mean surface air temperature, surface solar radiation, relative humidity, precipitation, wind speed, and sea-level pressure data observed at the Seoul weather station (SWS) for 2005–2015 were obtained from the KMA data portal (http://data.kma.go.kr). The SWS is the sole synoptic station in the region and meteorological conditions at the SWS are representative of those in Seoul (Ghim *et al.*, 2001; Yi *et al.*, 2016). We used the deviation of atmospheric variables from their own 30-year monthly averages, namely anomalies against monthly climatology, for calculating daily mean values for atmospheric data. Since the effects of atmospheric variables on concentrations of PMs vary from season to season, one year is divided into spring (March–April–May, MAM), summer (June–July–August, JJA), autumn (September–October–November, SON), and winter (December–January–February, DJF).

The Student's *t*-test was performed at a 95% confidence level to compare the values and linear trends of PMs concentrations and atmospheric variables. Least-squares linear regression analysis was used between daily $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations.

RESULTS AND DISCUSSION

Ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$

To check the characteristics of air quality in Seoul in the long term, a time series of the daily ratios of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ concentrations was examined for 2005–2015 (Fig. 1). The ratios fluctuate over time, resulting in its annual-mean (1.20) and daily standard deviation (0.39); the mean concentration of $PM_{2.5}$ is 20% higher than that of $PM_{2.5\text{-}10}$. This annual-mean ratio is lower than those in China and Taiwan: 2.23 in Beijing (Zhou *et al.*, 2015) and 1.33 in the downtown area of southern Taiwan (Lu *et al.*, 2016). However, the annual-mean ratio is higher than that

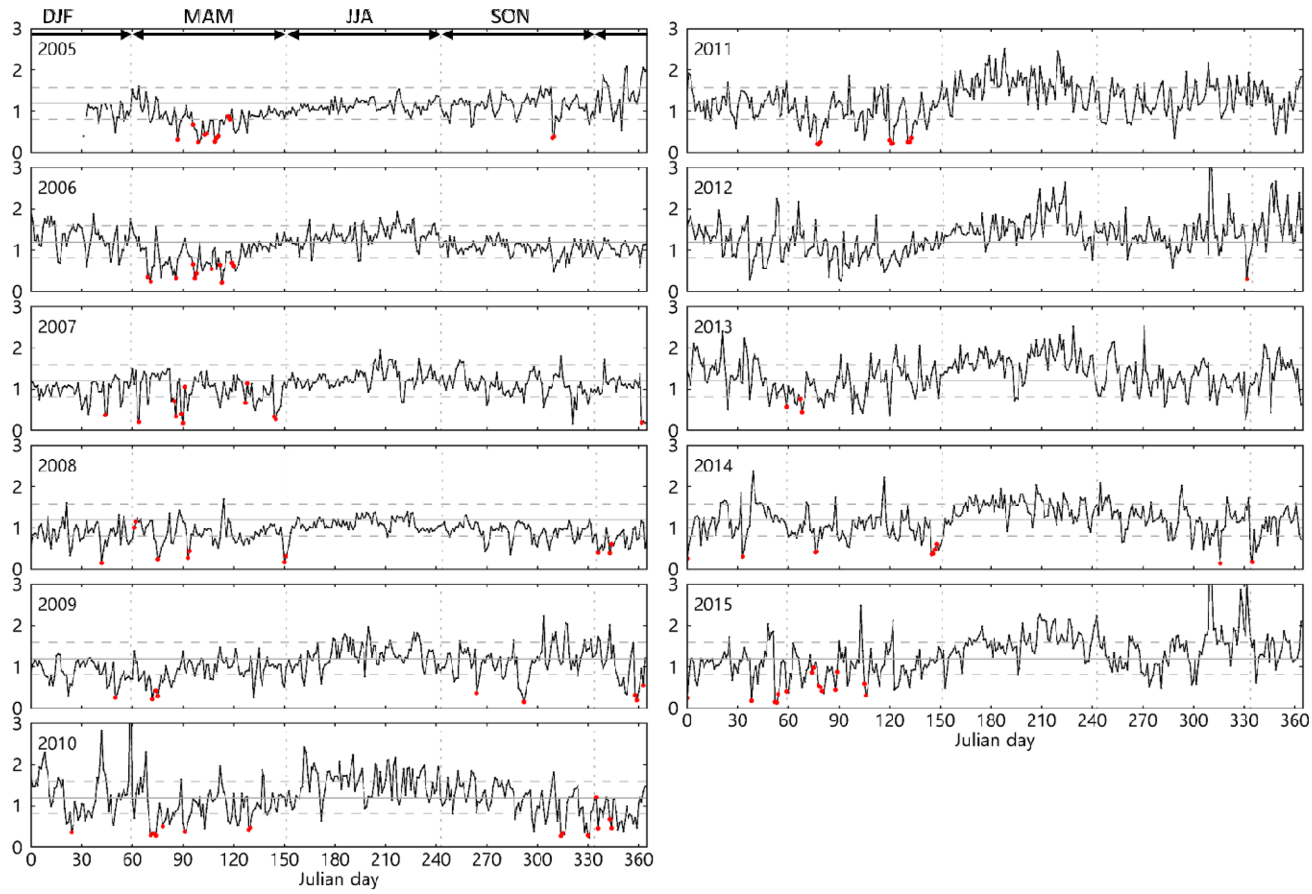

Fig. 1. Time series of the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ concentrations for the period 2005–2015. Red dots denote Asian dust days. Solid and dashed gray lines denote mean (1.20) and ± standard deviation (1.59 and 0.81), respectively. Dotted gray lines denote March 1, June 1, September 1, and December 1.

in other Asian regions, such as Hong Kong, the Philippines, Vietnam, and Japan where the ratio is around 0.82 (Cohen *et al.*, 2002). The fluctuation of the ratios indicates that $PM_{2.5}$ and $PM_{2.5\text{-}10}$ have different sources, processes, and emitting periods (Chen *et al.*, 1999; Co *et al.*, 2014). Dividing by season, the ratios in summer and winter are higher than those in spring and autumn (the p-value of the t-test, $p = 0.00$). The seasonal changes in the ratio are rather different from the results from Zhou *et al.* (2015) and Lu *et al.* (2016); they reported that the ratios in Beijing and southern Taiwan were high in autumn and winter and low in spring and summer, implying regional discrepancy in seasonal $PM_{2.5}$ proportion. In contrast, the ratios during Asian dust episodes (see red dots in the figure), mainly from the deserts in China and Mongolia, show relatively low values, with an average of 0.43.

To focus on the days when the emissions by combustion of fossil fuels and/or the secondary formations of $PM_{2.5}$ were predominant, we chose a value of one sigma above the annual mean ratio (i.e., 1.59 = 1.20 + 0.39) as a threshold, defined *the days with a high $PM_{2.5}$ proportion* as the days when the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ exceeded the threshold, and examined annual variations of the number of days with a high $PM_{2.5}$ proportion (Table 1). As seen in Fig. 1 and Table 1, the number of days is small in the 2000s, and large in the 2010s: The number of days per year in the 2010s (525 days/6 years) is 5.2 times larger than that in the 2000s (84 days/5 years). The ratios in the 2010s are also higher than those in the 2000s ($p = 0.00$): The average is 1.87 in the 2010s versus 1.75 in the 2000s. These results suggest that not only did the days with a high $PM_{2.5}$ proportion increase in number, but the proportion of $PM_{2.5}$ also increased.

Table 1 shows the yearly frequency of a high $PM_{2.5}$ proportion for each season as well. The total number of days with a high $PM_{2.5}$ proportion is the largest in summer, which is associated with the substantial increase in the frequency in the 2010s. The number of days in summer ranges 34–57 days in the 2010s but 0–16 days in the 2000s; the number of days per year in the 2010s (273 days/6 years) is 5.8 times larger than that in the 2000s (39 days/5 years). The proportion of the number of days in summer among the four seasons is 52% (273 days/525 days) in the 2010s, which is higher than 46% (39 days/84 days) in the 2000s. In the 2010s, the number of days in summer is the largest among the four seasons, which implies to be linked to the secondary formations of $PM_{2.5}$ and/or lesser amounts of long-range transport of Asian dusts (Chun *et al.*, 2001; Song *et al.*, 2006), while the season with the largest number of days varies by year in the 2000s. In spring, autumn, and winter, the number of days is also larger in the 2010s (34, 107, and 111 days in spring, autumn, and winter, respectively) than in the 2000s (3, 17, 25 days in spring, autumn, and winter, respectively), though the number is much smaller than that in summer.

The results of the linear regression analysis (Table 2) are in reasonable agreement with those mentioned above. Among the calculated linear regression coefficients, the slopes are over 1 and the largest in summer, reflecting the higher amount of $PM_{2.5}$ than $PM_{2.5\text{-}10}$ and the highest proportion of $PM_{2.5}$ concentration. Comparing the slopes in the 2000s with those in the 2010s, it was found that the slope significantly increases the greatest in summer. Hence it is possible to hypothesize that the changing rates of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ could have been different, which will be examined in the next subsection.

Trends in $PM_{2.5}$, $PM_{2.5\text{-}10}$, and Surface Air Temperature

Fig. 2 describes annual mean concentrations of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ for each season in 2005–2015. Their mass concentrations are relatively low in summer and autumn compared to spring and winter ($p = 0.20$ for $PM_{2.5}$ and $p = 0.00$ for $PM_{2.5\text{-}10}$); however, it does not mean that these low values need less attention because the annual-mean values—18.17–31.23 µg m^{-3} for $PM_{2.5}$ and 11.76–27.53 µg m^{-3} for $PM_{2.5\text{-}10}$ in summer, and 17.93–26.65 µg m^{-3} for $PM_{2.5}$ and 14.24–26.61 µg m^{-3} for $PM_{2.5\text{-}10}$ in autumn—are still higher than the current limit of the World Health Organization for the annual average—10 µg m^{-3} for $PM_{2.5}$ and 20 µg m^{-3} for PM_{10} (therefore, 10 µg m^{-3} for $PM_{2.5\text{-}10}$) (World Health Organization, 2006). Ghim *et al.* (2015) reported that annual average of $PM_{2.5}$ concentration decreased, whereas that of $PM_{2.5\text{-}10}$ did not show meaningful changes in 2002–2008. In this study, extending the analysis period to 2005–2015,

Table 1. The number of days when the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ concentrations exceeded the threshold (= 1.59) in each year.

Year	The number of days				
	MAM	JJA	SON	DJF	Total
2005	1	0	2	9	12
2006	1	16	0	11	28
2007	0	8	4	1	13
2008	1	0	0	1	2
2009	0	15	11	3	29
2010	8	46	11	16	81
2011	4	53	21	11	89
2012	3	37	21	27	88
2013	11	57	11	28	107
2014	4	34	12	12	62
2015	4	46	31	17	98

Table 2. Least-squares linear regression coefficients of the relationship [$PM_{2.5}$ μg m^{-3}] = slope × [$PM_{2.5\text{-}10}$ μg m^{-3}] + intercept. R is correlation coefficient and standard error is of the estimated gradient.

	Slope	Intercept	R	P-value	Standard error	N (valid days)
Total	0.802	6.908	0.774	0.000	0.011	3875
MAM	0.610	9.217	0.712	0.000	0.020	936
JJA	1.146	3.669	0.894	0.000	0.018	1012
SON	0.978	2.994	0.834	0.000	0.021	992
DJF	0.836	7.111	0.735	0.000	0.025	935
2000s	0.810	5.710	0.791	0.000	0.015	1737
MAM	0.609	9.512	0.690	0.000	0.031	419
JJA	1.160	1.041	0.934	0.000	0.021	460
SON	0.996	1.470	0.871	0.000	0.027	451
DJF	0.770	6.830	0.718	0.000	0.037	407
2010s	0.833	7.101	0.750	0.000	0.016	2138
MAM	0.604	9.187	0.711	0.000	0.026	517
JJA	1.411	2.092	0.884	0.000	0.032	552
SON	1.025	3.148	0.775	0.000	0.036	541
DJF	1.001	4.981	0.778	0.000	0.035	528

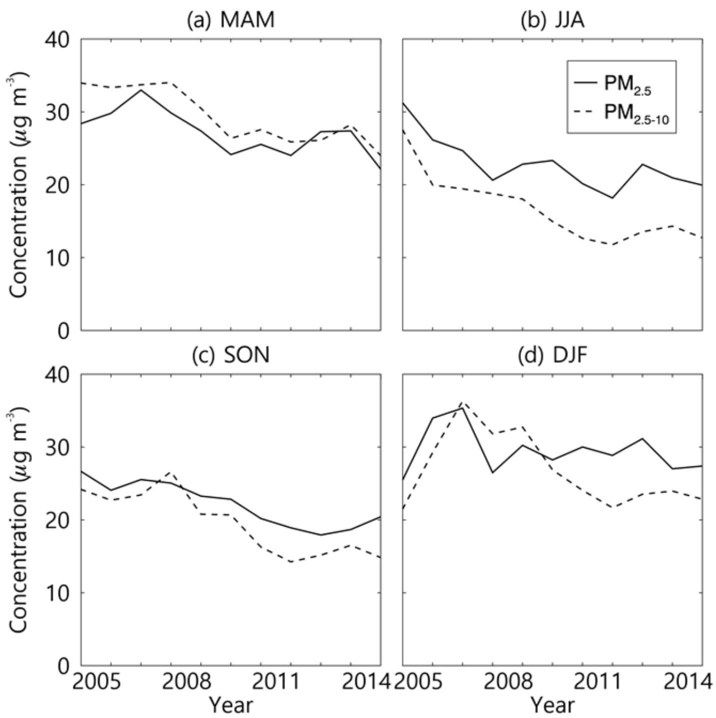

Fig. 2. The average concentrations of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ in MAM, JJA, SON, and DJF.

concentrations of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ are found to decrease significantly at the 95% confidence level in all seasons except for winter (Table 3). In summer (Fig. 2(b) and Table 3), concentration of $PM_{2.5}$ decreased much slower than that of $PM_{2.5\text{-}10}$: the difference of slopes (0.39 μg m^{-3} year^{-1}) is greater than their standard errors (0.24 μg m^{-3} year^{-1} for $PM_{2.5}$ and 0.23 μg m^{-3} year^{-1} for $PM_{2.5\text{-}10}$), meaning that the slopes of two PMs are significantly different ($p = 0.00$). The higher proportion (Fig. 1) and the slower decreasing tendency of $PM_{2.5}$ than those of $PM_{2.5\text{-}10}$ in summer should be noted. In spring and autumn, the differences of slopes (0.37 μg m^{-3} year^{-1} in spring and 0.32 μg m^{-3} year^{-1} in autumn) are also greater than their standard errors. In spring, however, air quality in Seoul is affected by even weak Asian dust. Thus, after excluding all Asian dust events, i.e., from two days before to two days after each Asian dust day, the difference of slopes (–0.77 μg m^{-3} year^{-1} for $PM_{2.5}$ minus –0.90 μg m^{-3} year^{-1} for $PM_{2.5\text{-}10}$ = 0.13 μg m^{-3} year^{-1}) becomes smaller than their standard errors (0.26 μg m^{-3} year^{-1} for $PM_{2.5}$ and 0.19 μg m^{-3} year^{-1} for $PM_{2.5\text{-}10}$), meaning that the slopes of two PMs are not significantly different. In autumn, further studies are needed to identify what caused the slow decrease of $PM_{2.5}$ because it was not explained by meteorological variables referred below.

The slower decreasing tendency in $PM_{2.5}$ compared to $PM_{2.5\text{-}10}$ in summer can be accounted for by the changes in

Table 3. The slope and standard error of regression of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations for the period 2005–2015. Bold denotes a slope significant at the 95% confidence level.

		MAM	JJA	SON	DJF
$PM_{2.5}$	Slope	**–0.651**	**–0.823**	**–0.826**	–0.240
	Standard error	0.225	0.239	0.134	0.300
	P-value	0.018	0.007	0.000	0.444
$PM_{2.5\text{-}10}$	Slope	**–1.021**	**–1.216**	**–1.142**	–0.744
	Standard error	0.172	0.234	0.214	0.441
	P-value	0.000	0.001	0.000	0.126

the emissions of air pollutants and/or the secondary formations of $PM_{2.5}$. The emissions of air pollutants in Seoul and surrounding metropolitan regions definitely decreased (Tables S2 and S3, Figs. S2 and S3), whereas concentrations of ionic components in dry and wet deposition showed increasing tendency (Tables S4 and S5). If other conditions that affect $PM_{2.5}$ concentration were specified as control variables, the decreases in the emissions of air pollutants, i.e., the precursors of secondary $PM_{2.5}$, would have induced the decreases in the secondary formations. Thus, it is inferred that changing atmospheric variables accelerated the secondary formations of $PM_{2.5}$. Wen *et al.* (2016) and Cao *et al.* (2017) showed that sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) values are the highest in summer over northern China due to the conversion-favoring factors such as high surface air temperature and relative humidity. Higher SOR and NOR values suggest more secondary particles produced by oxidation of gaseous precursors, implying the possible relationship between the long-term change of surface air temperature/relative humidity and secondary formations.

Annual averages of mean (T_{mean}), maximum (T_{max}), and minimum (T_{min}) surface air temperature anomalies for each season in 2005–2015 are shown in Fig. 3. In summer, all three surface air temperatures increased significantly at the 95% confidence level (Table 4), whereas there are no apparent increasing/decreasing trends for the other three seasons. This temperature increase is not likely to be induced by stronger solar insolation because the surface solar radiation observed at the SWS does not show any meaningful changes during summer (Table 5). According to Yeo *et al.* (2017), the increasing trend in Korean surface air temperature during summer is accounted for by the global-scale warming trend manifested in East Asia, whereas the variability is regulated by the atmospheric circulations over East Asia and the atmospheric teleconnections induced by the Pacific sea-surface temperature forcing. Therefore, it can be inferred that the temperature increase demonstrated in this study can be in part influenced by the global-scale trend.

Relationship between Trends in PMs and Surface Air Temperature

Since the slope of T_{max} is significantly larger than those of T_{mean} and T_{min} in summer: the values obtained by subtracting the slopes of T_{mean} and T_{min} from that of T_{max}—0.06°C year^{-1} and 0.10°C year^{-1}, respectively—are greater than their standard errors (p = 0.01 and 0.00, respectively), the diurnal variation of the slopes of hourly surface air temperature was examined. The increasing rates are only significant (p = 0.00–0.03) from 9 a.m. to 8 p.m. local time and not during the rest of the time in summer (c.f., the decreasing rates of two PMs are significant, p = 0.00–0.03 for $PM_{2.5}$ and p = 0.00 for $PM_{2.5\text{-}10}$, for every hour in summer). Fig. 4 shows a least-squares regression fit between the slopes of surface air temperature and PMs concentrations from 9 a.m. to 8 p.m. local time (12 data) in summer for the analysis period. Similar to Fig. 2, the decreasing rate of $PM_{2.5}$ is smaller than that of $PM_{2.5\text{-}10}$ (p = 0.04) during the daytime. The decreasing rates of $PM_{2.5}$ became smaller especially in the afternoon when the increasing rates of surface air temperature were large. The slope of regression fit between the slopes of surface air temperature and $PM_{2.5}$ is 1.74 times larger than that of $PM_{2.5\text{-}10}$. These results indicate that in summer, the gradual decrease of $PM_{2.5}$ during the daytime is closely related to high surface air temperature and its large increasing rate. Furthermore, considering the emissions of air pollutants, dry and wet deposition, and that air temperature accelerates the secondary formations, it seems proper to explain that the gradual decrease in $PM_{2.5}$ can result from the increased secondary formations induced by the increase in surface air temperature. Our results suggest that, even when PMs concentrations decrease as a result of the implementation of air pollution reduction policies, $PM_{2.5}$ can decrease more slowly than $PM_{2.5\text{-}10}$ under changing atmospheric conditions; therefore, the proportion of $PM_{2.5}$ becomes high.

Among other atmospheric variables related to concentrations of PMs (Table 5), relative humidity in summer can also contribute to the increase of the secondary formations of $PM_{2.5}$ (Wang *et al.*, 2005). In Seoul, however, mean relative humidity in summer is 70.9% without showing significant changes during the analysis period. In addition, as referred in the Introduction, rain and wind can lower PMs concentrations by the scavenging effect and the dilution effect, respectively. In terms of precipitation, coarser particles are more effectively washed out than fine particles under the same rainfall intensity (Hu *et al.*, 2006; Tiwari *et al.*, 2012; Olszowski, 2015). However, there was no significant change in precipitation in summer. We also examined the changes in light rainfall—from 0 mm day^{-1} to 1–10 mm day^{-1}—and the number of days with light rain for the analysis period, but they did not show any significant change, either. Wind speed, on the other hand, increased significantly in all seasons, which can be a cause of decreased PMs. However, it is difficult to separate the influence of wind speed on $PM_{2.5}$ and $PM_{2.5\text{-}10}$. Because

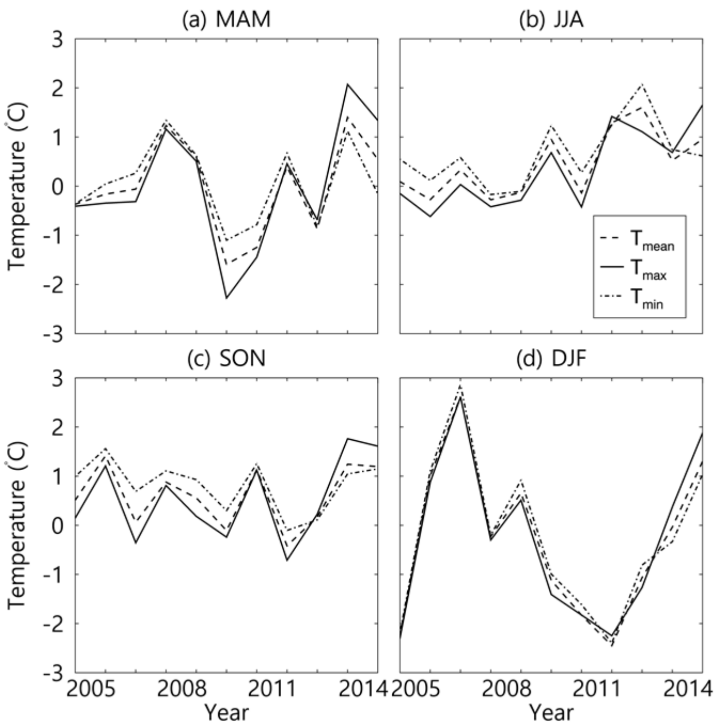

Fig. 3. The average of anomalous T_{mean}, T_{max}, and T_{min} in MAM, JJA, SON, and DJF.

Table 4. The slope and standard error of regression of anomalous T_{mean}, T_{max}, and T_{min} for the period 2005–2015. Bold denotes a slope significant at the 95% confidence level.

		MAM	JJA	SON	DJF
T_{mean}	Slope	0.044	**0.131**	0.009	–0.040
	Standard error	0.096	0.050	0.062	0.161
	P-value	0.656	0.028	0.883	0.809
T_{max}	Slope	0.127	**0.191**	0.083	0.009
	Standard error	0.120	0.050	0.078	0.168
	P-value	0.319	0.004	0.315	0.959
T_{min}	Slope	–0.006	0.096	–0.047	–0.069
	Standard error	0.081	0.059	0.049	0.159
	P-value	0.939	0.137	0.372	0.677

Table 5. The slope and standard error of regression of anomalous surface solar radiation, relative humidity, precipitation, wind speed, and sea level pressure for the period 2005–2015. Bold denotes a slope significant at the 95% confidence level.

		MAM	JJA	SON	DJF
Surface solar radiation	Slope	–0.001	0.063	0.015	–0.043
(MJ m^{-2})	Standard error	0.094	0.097	0.066	0.045
	P-value	0.990	0.532	0.828	0.358
Relative humidity	Slope	–0.116	–0.326	0.004	0.228
(%)	Standard error	0.285	0.262	0.225	0.335
	P-value	0.693	0.246	0.987	0.514
Precipitation	Slope	–7.680	–25.001	–1.649	2.007
(mm)	Standard error	4.177	33.664	16.624	2.299
	P-value	0.099	0.477	0.923	0.405
Wind speed	Slope	**0.052**	**0.042**	**0.038**	**0.031**
(m s^{-1})	Standard error	0.013	0.015	0.010	0.013
	P-value	0.003	0.018	0.005	0.044
Sea level pressure	Slope	0.108	–0.021	–0.020	0.010
(hPa)	Standard error	0.081	0.074	0.052	0.126
	P-value	0.218	0.786	0.708	0.937

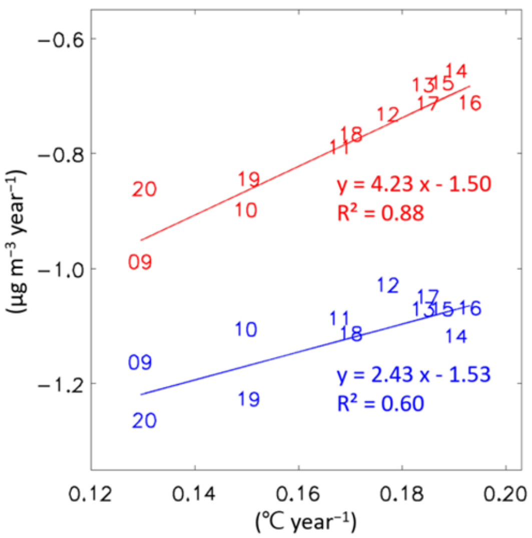

Fig. 4. A least-squares regression fit between the slopes of surface air temperature and the slopes of $PM_{2.5}$ (red) and $PM_{2.5\text{-}10}$ (blue) concentrations, respectively, from 09 LST to 20 LST (12 data) in JJA for the period 2005–2015. Numbers (09 to 20) represent local time.

high wind speed can also result in high PMs concentrations due to the resuspension and long-range transport of PMs under well-dispersed conditions (i.e., high mixing-layer height), as well as low PMs concentrations due to the dilution effect (Cheng and Li, 2010), and because dust resuspension may not increase concentrations of both $PM_{2.5}$ and $PM_{2.5\text{-}10}$ (Zhang *et al.*, 2018), further studies are needed to identify the long-term relationship between wind speed and two PMs separately. Finally, sea-level pressure, related to stabilization/destabilization of atmosphere and thus accumulation/dilution of PMs, did not show any significant change in summer.

SUMMARY

We analyzed $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations to examine the differences between their long-term changes in Seoul, South Korea. For the period 2005–2015, the number of days with a high $PM_{2.5}$ proportion increased in summer, implying different changes in the $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations. The decreasing rate of the $PM_{2.5}$ concentration was significantly smaller than that of the $PM_{2.5\text{-}10}$ concentration during summer when the surface air temperature increased significantly. Moreover, the decreasing rate of the $PM_{2.5}$ became low during the daytime, especially in the afternoon, when the increasing rate of the surface air temperature became high. Considering the mechanisms of secondary formation, these results suggest that the long-term increase in surface air temperature can induce the increase of secondary $PM_{2.5}$ formation and, consequently, the slower decrease of $PM_{2.5}$ compared to $PM_{2.5\text{-}10}$. According to our results, even when concentrations of PMs decrease as a result of the implementation of air pollution reduction policies, $PM_{2.5}$ can decrease more gradually than $PM_{2.5\text{-}10}$ due to changes in the atmospheric conditions; therefore, the ratio of $PM_{2.5}$ to $PM_{2.5\text{-}10}$ becomes high.

$PM_{2.5}$ is formed not only from anthropogenic emissions but also from secondary formation, and various meteorological variables can influence the mechanisms of formation by season. In this study, by examining both the $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations and the meteorological variables, we deduced the effects of increased surface air temperatures on the decrease of the $PM_{2.5}$ concentration in summer. There are still uncertainties in this study: (1) Errors in the measurement of $PM_{2.5}$, PM_{10}, and the surface air temperature exist; (2) the number of sites where $PM_{2.5}$ was measured is not constant; (3) emission data of air pollutants are available only on an annual basis; and (4) it is difficult to thoroughly prove a causal relationship between the $PM_{2.5}$ concentration and the surface air temperature with only observation data. Nonetheless, our results emphasize the necessity of monitoring both long-term PM concentrations and meteorological variables in responding to future climate change and the subsequent air quality change. The findings in this study will be useful for providing a basis for the independent and separate establishment of reduction policies for $PM_{2.5}$ and $PM_{2.5\text{-}10}$.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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